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RADIATION SURVEILLANCE OF THE ENVIRONMENT

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Cincinnati, Ohio

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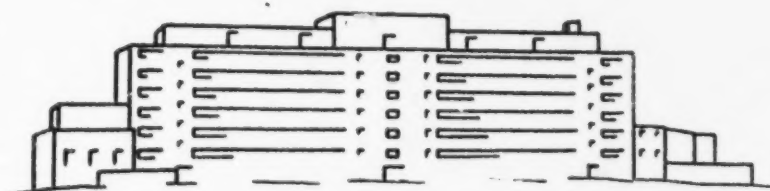
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U.S. DEPARTMENT OF HEALTH,
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Public Health Service

RADIATION SURVEILLANCE OF THE ENVIRONMENT

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by

Conrad P. Straub

Introduction

The continued development and use of nuclear energy is resulting in the release of a wide variety of radioactive materials to the environment. In the past, naturally occurring radionuclides and cosmic radiation were essentially the sole sources of such contamination. At present, however, radioactive contamination can result from multiple sources including uranium and thorium ore processing plants; fuel fabrication facilities; reactor installations - both fixed and mobile; fuel reprocessing plants; beneficial applications of radioisotopes in medicine, research, and industry; and fallout from nuclear tests.

Release of radioactive materials from any of these operations can lead to the direct contamination of the occupational worker, the general population, or various segments of the environment. Although the responsibility for control of such pollutants is with the operator of the facility involved, particularly as it relates to the exposure of the worker during normal and emergency operation and the general population during accidental release of radioactive materials, local and national health agencies are necessarily interested in the widescale public health implications of such releases. Health agencies have another interest, and that stems from the medical uses of radiation in all its forms for diagnosis and therapy.

The purpose of this paper is to indicate the variety of tests required for a detailed appraisal of the public health implications of the planned or accidental release of radioactive materials to the environment.

Maximum Permissible Exposure

Our present measure of gauging significance of contamination of the environment is in relation to the maximum permissible concentration (MPC) values recommended by the International Commission on Radiological Protection for many radionuclides in water and air (1). Recently, it has been suggested that the MPC values for water are equally applicable to food substances following a change in units from $\mu\text{c/ml}$ to $\mu\text{c/gm}$ wet weight (2). Furthermore, the permissible occupational levels (1) have been reduced by a factor ranging from 10 to 100 to apply to the general population because of the potential genetic effects arising from the radiation exposure of large segments of the population. The MPCs were calculated on the basis of continuous intake of, or exposure to, the radionuclide in question for long periods of time ranging from 40 to 70 years.

Before the extent of contamination may be determined, a system of monitoring stations must be established to measure the levels of radioactivity in the environmental media of interest and to note changes taking place in these levels. Two approaches may be used to environmental monitoring or surveillance - gross activity or specific radionuclide analyses. Measurement of gross

activity will serve as a basis for routine control in nuclear energy establishments and to indicate accidental release of radioactivity in plant operations. It will also be helpful in following the movement of radioactive clouds resulting from nuclear detonations when such detonations take place in the open. Such measurements will be useful also in estimating the external dose from such releases.

If one wishes to determine internal radiation hazard, information is needed on the specific radionuclides present. This is due to the wide difference in response of body tissues to specific chemical elements and, consequently, their radionuclides.

Since the required analyses are more or less a function of the sources of radioactivity, the remaining portions of this paper will be concerned with this topic. The suggested analytical tests are reported in terms of the various operations listed in the initial paragraph of this paper. No consideration is given to body assay techniques, since this is the subject of a companion paper prepared by Dr. C. G. Stewart.

Ore Production and Processing

In mining uranium, as in any mining operation, care must be taken to protect the workers from exposure to dusts, and from prolonged contact with the ore (radon inhalation). There will be some losses of material in transport and during storage, but the level of radioactivity in most cases will be low. Gross alpha or beta-gamma

measurements with suitable instrumentation will provide information on potential external radiation hazards. Analyses for radon will indicate the extent of exposure to this source. Where uranium ores are stored in the open, it may be desirable to check nearby streams for levels of soluble radium which may have been leached from these accumulations during storage. Vegetation and soil in the area may be checked for radium, since this is the most hazardous radionuclide.

During processing of the ores for uranium recovery, liquid wastes may be discharged which contain uranium, radium, and the daughter products of these elements. In a few cases, the tailings are discharged directly into the stream, but in most instances they are stored and the wastes discharged to tailings ponds which permit settling of most suspended matter. The tailings ponds discharge into nearby water courses. Experience has shown that radium levels in streams below these plants are somewhat higher than those up-stream. It is necessary, therefore, to measure the radium levels in water above and below the plant, as well as to measure the activity in the bottom muds and the associated stream biota. Analysis of muds below uranium processing plants is a well known procedure for control of such operations. Where these stream waters are used for irrigation, plant uptake may be of consequence and analyses should be made for the removal of radium and uranium by soil and vegetation which serve as sources of animal and human food.

In all of these cases, useful data may be obtained by the measurement of gross activity associated with particulate matter.

Fuel Production

The processing of prepared uranium ore for the production of its brown oxide (UO_2), its green salt (UF_4), or the gaseous UF_6 presents the ordinary problems associated with chemical industries using toxic solvents and extracting solutions and complicated reactors. Mists and fumes must be controlled and contaminants removed by scrubbing, filtration, or other means to prevent atmospheric contamination.

Refined uranium metal, an end product of chemical processing, is fabricated for reactor fuel. During fabrication, which includes such operations as rolling, extruding, machining, and other metallurgical treatments, fumes of uranium oxide may be given off as atmospheric contaminants. Water used in cooling or clean-up contains uranium dusts and chips, but these can be removed easily and will not contaminate any waterway or sewer system.

Environmental contamination may be controlled by minimizing the release of atmospheric contaminants and by providing facilities for the removal of uranium dusts and chips from liquid wastes prior to their discharge.

Reactor Installations

Most older reactors are of the single flow through coolant type, the coolants being air or water. As a result, relatively large volumes of contaminated air or water are discharged.

To minimize the formation of induced activity, the coolant is pre-treated to remove most particulate matter. Before release to the environment, the air is filtered to remove particulate activity and the water is stored to permit decay of the short-lived radionuclides formed in or picked up in passing through the reactor. Power and many experimental reactors generally operate on a closed coolant cycle with passage of the coolant through ion exchange columns or other treatment devices for the removal of radioactivity.

With proper operation, the amounts of radioactivity discharged from single pass- or recirculating-type reactors should be quite small, and will be at or well below the MPC values. Local conditions will govern the release of radionuclides.

Coolant air, after passing through the reactor and treatment to remove particulate matter, is usually discharged through stacks for dispersion of the argon-41 produced. Monitoring may include analyses to determine the levels of this radionuclide during dispersion under various meteorological conditions.

Gross or radionuclide activity may be measured prior to or during release of wastes into streams, and will include measurement of radioactivity in reactor coolant water (where single-pass cooling is employed as at Hanford), fuel element canal storage water, wastes from laboratory areas and laundries, and washings from decontamination pads or cells. Because of deposition, adsorption, and/or ion exchange in the stream environment, bottom sediments must be examined. Similarly, because of adsorption and metabolism, there is a need for

determining the levels of accumulation in plankton, fish, vegetation, and animals.

This type of monitoring has resulted in the voluntary reduction of maximum permissible levels of discharge into the stream environment by Hanford of the following radionuclides (3): Ru-106 from 0.1 to 10^{-3} $\mu\text{c/ml}$ (the ICRP values (1) for this nuclide in water are given as 0.1 $\mu\text{c/ml}$ for the kidney and 10^{-4} $\mu\text{c/ml}$ for the GI tract), P-32 from 2×10^{-4} to 3×10^{-7} $\mu\text{c/ml}$, Na-24 from 8×10^{-3} to 2×10^{-6} $\mu\text{c/ml}$, and H-3 from 0.2 to 0.05 $\mu\text{c/ml}$. From this it is obvious that continuous surveillance is needed to determine permissible levels of discharge as a function of downstream water use.

Where water is used for irrigation, the various soil types should be sampled to determine exchange and adsorption of specific radionuclides, and analyses should be made of the crops growing in this area to measure uptake. This is taking on more significance in view of recent findings of higher uptake by plants from the water environment than from the soil environment.

With the advent of mobile reactors, special provisions will have to be made for analyses of estuarine and marine waters.

In the case of reactor failures or accidents, the analyses required will be similar to those indicated for fuel reprocessing plants. The comments relating to the release of iodine during chemical reprocessing of fuel elements apply equally well to the case of reactor fuel failures as was indicated during the Windscale reactor accident. Early monitoring and control involved the determination

of iodine in a variety of environmental media.

Fuel Reprocessing Plants

When a spent reactor fuel element is dissolved in HNO_3 or other medium at the beginning of chemical processing for the recovery of the unfissioned fuel, those fission products that are gaseous under the conditions employed - chiefly iodine and the inert gases krypton and xenon - will be released in the off-gas stream. If fuel elements are cooled for short periods after removal from the reactor and prior to dissolution, large quantities of essentially pure isotopes of iodine will be released (4). Unless iodine is very thoroughly scrubbed from the gas stream before release to the stack, the iodine will become a distinct radiological hazard to people working at short diffusion distances from the stack both inside and outside the plant area.

The iodine released, being in either gaseous or particulate form, may be inhaled or pose an ingestion hazard due to deposition on vegetation and consumption by animals or man. Extensive monitoring of vegetation and animal thyroids is carried out in the vicinity of fuel reprocessing plants to maintain surveillance over this particular radionuclide. It is for this reason that there is such widespread interest in a field-type iodine sampler which will collect and detect iodine at low concentration, will be low in cost to encourage installation at a large number of sampling stations, and will be used only once and discarded since decontamination will be difficult if not impractical (4).

Milk obtained from cattle feeding in the vicinity of chemical reprocessing plants releasing radioiodine may show substantial levels of this radionuclide.

Again citing the experiences of Hanford, the suggestion has been made of reducing the MPC value for I-131 in air from 3×10^{-9} to 3×10^{-13} $\mu\text{c}/\text{ml}$ and H-3 from 2×10^{-5} to 3×10^{-6} $\mu\text{c}/\text{ml}$. They have also introduced a level for I-131 in vegetation equal to 1×10^{-5} $\mu\text{c}/\text{g}$ (3).

Spent Radionuclides

Radionuclides are widely used in hospitals for diagnosis and therapy as well as medical research. In diagnosis and therapy they may be used in gaseous, liquid, or solid form, and may be taken internally or used externally as solid sources. Radioactivity, in the form of spent solutions or body discharges, may have to be released. Generally, because the materials are of relatively short half-life, they may be stored for decay or diluted and discharged into the hospital sewerage system. On the basis of the quantities discharged at present, the latter method poses no particular environmental hazard. Obviously, solid sources will not be disposed of indiscriminately, but will be returned to the manufacturer or to the atomic energy authority for disposal.

In medical and biological research, in addition to liquid wastes, we have the problem of solid waste disposal including such materials as straw bedding, cage cleanings, and animal carcasses. These are disposed of generally by incineration. The waste gases which result

should be monitored along with the residual ash. The latter is packaged prior to disposal by burial. Gross activity measurements should suffice for most hospital operations.

Laundrying of contaminated clothing and other articles should take place after monitoring of the material to determine its radiation level, or special facilities and techniques should be provided.

Similar comments would relate to the release of radioactive materials following research in industry or agriculture. The variety of radionuclides released would be greater. Long-lived radionuclides should not be released into the environment, but should be stored for reduction to a solid state and incorporation in some medium like concrete before disposal in suitable containers by burial on land or in the ocean.

Fallout

Nuclear tests have resulted in the widespread release of radioactive materials, some of which are deposited close to their site of formation; others such as Sr-90 and Cs-137 are carried into the stratosphere from which they may be deposited slowly over large areas distant from the point of detonation.

Extensive surveillance operations, both on-site and off-site, have been carried on for several years. These operations include the measurement of gross activity in air, rainfall, and soil, and the determination of specific radionuclides. Numerous studies have been conducted to measure the uptake of Sr-90 and Cs-137 from fallout

through the various elements of the food chain to man. The analyses include determination of activity in air, rainfall, surface waters, soil, vegetation, food materials such as milk, fish, shellfish, etc., human and animal bone, and other tissues.

Fallout is also measured in relation to normal background (natural radioactivity) associated with atomic plant operations. Thus, to measure the real contribution of radioactivity to the environment from such operations, the amount contributed by fallout must be known and corrections made for it. In many instances, it is difficult to determine the level of activity discharged from some of our nuclear plants because of the masking effect of fallout.

Miscellaneous

In the preceding discussions little mention was made of solid waste disposal and the analytical controls associated with such operations. Generally, monitoring consists of measuring external radiation levels and the activity of ground waters traversing the burial site. In some instances, vegetation, notably trees and leaves, may be better indicators of the release of activity from such operations.

Nothing was said earlier about the release of activity into the seas or oceans. Discharges may be direct, as at Windscale, and involve low level wastes, or they may involve the release of packaged materials some distance off shore. Specific analyses may be required depending upon local conditions. For example, monitoring at Windscale includes sea water, fish nets and other fishing paraphernalia, sea sand, fish, and a specific kind of sea-weed. In the case of packaged disposal,

as long as the integrity of the container is maintained, there should be no release of radioactivity. Generally too, such material is prepared into a monolith which should erode slowly and consequently there should be a very slow release of radioactivity.

The total body counter, or similar types of devices, for the measurement of large samples should find extensive use as a scanning device to more extensive analyses. This device has had its greatest recent development and application in measuring total body activity, but it has been used also to determine the radioactive contamination of foodstuffs (5).

Samples generally will be collected in the plant area and immediate environs by the operator of the plant. The number, kind, and frequency of sample collections and analyses vary with the type of installation, its location, and method of operation. Gross determinations, particularly of air and water, may be made on a daily basis and sample aliquots composited for radionuclide identification on a monthly basis.

In focussing our attention on the monitoring of radioactive materials in waste streams, we must not lose sight of the fact that some stable materials associated with these discharges may also be toxic. Examples include mercury, solvents, nitrates in high concentration, and high fluoride levels.

To assist in assessing changes in a given environment due to the siting of a nuclear facility, pre-operational monitoring of the air, water, soil, vegetation, and animals in the area is desirable to establish so-called background levels. Such data should be obtained over a period of one or two years prior to plant start up to define seasonal variations in radioactivity levels.

Summary

The analytical methods used for radiation surveillance have been developed over a period of years by highly competent radiochemists using the latest instrumentation. In arriving at the kinds of analyses to be made for public health control, the competency of the staffs involved and availability of equipment and instrumentation will have to be considered. At the present time, the trend is toward instrumental identification of many radionuclides, particularly the gamma emitters. This trend will increase and should simplify analyses and encourage the taking of more samples for a better definition of what is happening in the environment.

The analytical tests required to define environmental contamination from each of the sources discussed in the text are summarized below:

Ore production and processing - gross alpha and beta-gamma determinations of particulate matter collected in air samples, and specific analyses for radon and radium in the case of uranium mining and processing and analogous members in the case of thorium mining and processing. Gross alpha and beta-gamma determinations of waste discharges and

levels of radium in bottom muds, vegetation, plankton, and water.

Fuel production - air monitoring for gross alpha and beta-gamma particulate activity and gross radioactivity measurements of liquid discharges.

Reactor installations - normal operation - air and water monitoring for gross alpha and beta-gamma activity on a continuous basis, followed by radionuclide analyses of liquid discharges on monthly composite samples. Analyses of various environmental media below installations will be required. Monitoring of solid wastes will be necessary. Analyses of sea water and its associated environment in case of ocean discharges will be required.

Reactor installations - emergency operations - air monitoring for gross activity and radionuclide analyses for iodine and other particulates. In some instances analyses for tritium may be necessary.

Fuel reprocessing - monitoring of stack effluents for gaseous and volatile fission products released during reprocessing. Monitoring by checking vegetation and animal thyroids for iodine-131 is of value. Following removal of long-lived components and storage for decay, residual wastes discharged into the environment should be monitored. Analyses will determine the fate of these materials in the environment.

Spent radionuclides - monitoring of air and sewage to see that the MPC values are not exceeded.

Fallout - monitoring of air, rainfall, soil, plant life, etc., for gross activity and radionuclide analyses for strontium-90 cesium-137, and others in milk, foodstuffs, soil, human and animal bones, etc.

Conclusions

Radiation monitoring of all elements of the environment is needed to evaluate current radiation exposure to man from these sources and to relate current findings to MPC values for continuous long-term intake. Some studies, notably those at Hanford, have pointed to the need for extending surveillance beyond the property line and the discharge outlet. Discharge at sub-MPC levels may become necessary because of biological assimilation and uptake. Downstream water uses may also exert some influence on permissible levels of discharge. Continued radionuclide monitoring of the environment will provide data on the adequacy of currently recommended MPC values.

Specific precautions on taking samples have not been discussed, because these can be covered most satisfactorily during consideration of sampling and analytical techniques.

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